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USE OF TUNGSTEN-184 AS A THERMAL FLUX MONITOR AT HIGH TEMPERATURES

by Lauren L. Ball, Paul J. Richardson, and Dean W. Sheibley

Lewis Research Center
Cleveland, Ohio

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ABSTRACT

Tungsten-184 has been found to be a useful dosimeter for thermal neutrons, particularly for high-temperature applications. Correlation of neutron flux with cobalt-59 activation and uranium-235 fission density for short-term irradiations is shown. The tungsten-184 thermal-neutron capture cross section in the Plum Brook Reactor was found to be 1.94±0.08 barns. Cadmium ratios for tungsten-184 are about one-fifth of the value found for cobalt-59. The radiochemistry and counting techniques for tungsten-185 are described.

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SUMMARY

Tungsten-184 is a useful dosimeter for thermal neutrons, particularly for high-temperature applications. Correlation of neutron flux with cobalt-59 activation and uranium-235 fission density for short-term irradiations is shown. The tungsten-184 thermal neutron capture cross section in the Plum Brook Reactor was found to be 1.94±0.08 barns. Cadmium radios for tungsten-184 are about one-fifth of the value found for cobalt-59. The radiochemistry and counting techniques for tungsten-185 are described.

INTRODUCTION

Short-term irradiations (<40 hr) of tungsten-clad, tungsten - uranium dioxide fuel plate test specimens at high temperatures produced neutron dosimetry problems. The high temperatures of the irradiations (2500 to 3100 K) did not permit the use of standard dosimeters such as cobalt, dysprosium, or gold to determine the thermal flux. (The melting points of cobalt, gold, and dysprosium are 1768, 1336, and 1680 K, respectively, in contrast to the melting point of tungsten, 3683 K.) The use of fission products such as cesium-137 or zirconium-95 as a measure of burnup was not reliable since specimens lost fission products during irradiation. The shorter term irradiations, in some cases 4 hours or less, did not produce sufficient change in the uranium-236 to uranium-235 ratio (U^{236}/U^{235}) to accurately determine the uranium burnup. Hence, the thermal flux which the specimens saw could not be determined accurately.

The dosimetry problem was resolved by using naturally occurring tungsten-184 in the specimen cladding as the thermal flux monitor. Tungsten-185 activity, which is produced by neutron activation of tungsten-184, was used to determine the thermal neutron flux.

The thermal cross section of tungsten-184 has been determined by several investigators. The recommended value is 1.8±0.2 barns (ref. 1). Values as low as 1.65 barns (ref. 2) and as high as 2.28 barns (ref. 3) have been reported. Due to the variations in the cross section of tungsten-184 in the literature, it was necessary to determine its effective thermal cross section in this application.

The radiochemistry of tungsten has been covered by Mullins and Leddicote (ref. 4). However, since tungsten-184 is a β emitter, it was necessary to refine the dissolution methods, the decontamination methods, and develop counting techniques to ensure the accuracy of using tungsten-184 as a flux monitor.

EXPERIMENTAL DETAILS

The fueled test specimens for which this dosimeter was developed were 2.54 by 2.54 by 0.127 centimeter thick. The fuel was enriched uranium oxide in a matrix of metallic tungsten. All test plates were clad with ~0.013 centimeter of tungsten and encased in a picture frame of tungsten. Each fueled test plate was suspended inside a stainless-steel capsule and irradiated in the HT-1 test hole of the Plum Brook Reactor (PBR). The actual thermal-neutron fluxes which the specimens saw varied from 10^{13} to 10^{14} neutrons per square centimeter per second depending on the position in the HT-1 facility. The length of irradiation varied from 1 to 40 hours. The HT-1 facility nuclear environment is described in the following table.

Gamma heating at 60 MW, W/g	~3
Thermal flux, N/(cm^2)(sec)	~1×10^{14}
Fast flux >0.1 MeV, N/(cm^2)(sec)	2×10^{13}

Two methods were used for dissolving the fuel test plates. If total uranium determination and burnup analysis were to be performed, the fuel plates were dissolved in an acid solution of two parts phosphoric acid (H_3PO_4) and one part perchloric acid ($HClO_4$). This method achieved complete dissolution of the tungsten and uranium oxide. But because of the high phosphate ion concentration, separation of the tungsten activities was difficult.

For determining the tungsten activities only, samples of the tungsten picture frame surrounding the fuel plate were removed. Each sample was then dissolved in a mixture of hydrofluoric (HF) and nitric acids (HNO_3). This method is rapid and does not dissolve the uranium oxide. Care was taken when removing the tungsten picture frame piece so that no fuel was included. This eliminated much of the fission product contamination.

Using opposite corners of the tungsten picture frame of the fuel test plates permitted us to determine whether a significant flux gradient existed across the specimens. Aliquots were taken from the solutions of the tungsten corners and 20 milligrams of tungsten

carrier were added. Tungsten was precipitated as tungstic acid ($H_2WO_4 \cdot XH_2O$) in acid. The precipitate was dissolved in ammonia solution (NH_4OH) and scavanged with iron several times. Additional specific decontaminations were performed as required. Interfering activities were detected using a gamma spectrometer. Tungstic acid was finally precipitated and ignited to tungsten trioxide (WO_3). The ignited WO_3 was filtered onto a standard 2.4-centimeter fiberglass filter. The filtered WO_3 was then mounted on a stainless-steel planchet. The precipitate was covered with Mylar film (0.0005 cm thick). The mounted samples were then β-counted in a windowless gas-flow proportional counter.

The conversion of counts to disintegrations was achieved by using the scattering and self-absorption (F_{SSA}) curve shown in figure 1. This curve was prepared by counting a

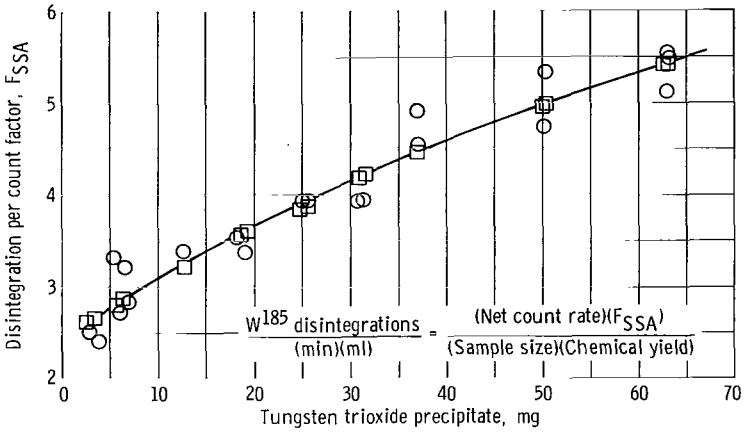

Figure 1. - Tungsten-185 counting efficiency curve. Absorption and self-scatter of 0.43 MeV β's in tungsten trioxide. Test area, 4.5 square centimeters of chimney mount; Mylar covered sample mount. To use: Obtain net count rate of Mylar mounted sample on windowless gas-flow proportional counter (geometry, ~50 percent). Determine disintegration-count - scattering and self-absorption factor from milligrams of tungsten trioxide precipitate.

known aliquot of carrier-free W^{185} tracer in a 4π counter. When the disintegration rate of W^{185} was established, samples containing varying weights of tungsten (3 to 65 mg W as WO_3) plus known amounts of W^{185} tracer were separated. The count rate on each precipitate was compared with the known disintegration rate. The disintegration per count d/c factor in figure 1 was established for each weight of WO_3 precipitate mounted in the specified manner. The working curve (fig. 1) is a least squares fit of the experimental data.

The precision of the radiochemical procedure is estimated to be ±2.5 percent. (All uncertainties in this report are at the 95-percent confidence level.)

DISCUSSION AND EXPERIMENTAL RESULTS

The recommended (ref. 1) 2200-meter-per-second cross section of W^{184} is 1.8±0.2 barns. Our comparison of W^{184} with Co^{59} (σ = 37.2 b) based on 13 activations in a thermalized reactor spectrum produced an effective thermal cross section of 1.93±0.05 barns. The comparison with U^{235} (σ_f = 522 b[1]) burnup at high temperatures based on five irradiations showed an effective thermal cross section of 1.96±0.36 barns. The effective cross section of W^{184} based on an unweighted average of all 18 values is 1.94±0.08 barns. The comparison of the cadmium ratios of cobalt and tungsten showed about a 9-percent epicadmium contribution to W^{184} activation based on similarity in the neutron spectra in HT-1 and RA-8 locations.

To check the response of W^{184} against Co^{59} at lower temperatures (<1500 K), 13 irradiations of varying lengths of time were made in the HT-1 facility of the Plum Brook Reactor (PBR) at different positions.

In each irradiation, a cobalt and a tungsten flux wire were placed together in a stainless-steel capsule and irradiated under the same low-temperature conditions as the fuel plates. The cobalt wires were removed and the Co^{60} activity determined. The tungsten wires were dissolved and the W^{185} activity determined. Based on a cobalt 2200-meter-per-second cross section of 37.2 barns, the effective thermal cross section of W^{184} is 1.93±0.05 barns.

Further confirmation of the effective thermal cross section of W^{184} was obtained in the temperature range 1100 to 3300 K by correlating the W^{185} activity found in the tungsten cladding with mass spectrometer burnup analysis of the uranium in the test specimens.

The five mass spectrometer values were calculated from the increase in the U^{236} content. When combined with the U^{235} capture-to-fission cross-section ratios α_5, these data may be used to determine the atom percent fissions in U^{235} (ref. 5). The fluence was then calculated from the effective U^{235} fission cross section (522 b) used for the PBR. Precision of the data in table I was limited by the low burnup and relatively small change in U^{236} content in these irradiations. And, if the preirradiation specimens analyzed for initial U^{236} content are not representative of the specimens irradiated, an indeterminate error in the burnup calculation will occur. This error is larger at lower burnups. There is another source of error in the value of α_5. This ratio is a function of neutron temperature and cadmium ratio (ref. 6). The specimens were small enough to ignore the neutron contributions from the specimens. Therefore, the value of α for a thermalized core, 0.175, was used. The uncertainty is estimated to be about 5 percent.

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[1]The fission cross section of U^{235} at the operating temperature of the Plum Brook Reactor has been calculated as 522 barns.

TABLE I. - CORRELATION OF MASS SPECTROMETER BURNUP

WITH TUNGSTEN-184 ACTIVATION

$$\left[F_5 = \frac{N_5^o\left(R_{6/5} - R_{6/5}^o\right)}{R_{6/5} + \alpha_5\left(1 + R_{6/5}\right)}\right.$$; α_5, capture to fission cross section ratio (U^{235}) N_5^o, initial atom percent abundance of U^{235}.]

Fuel specimen	Initial U^{236} to U^{235} ratio, $R_{6/5}^o$	Postirradiation U^{236} to U^{235} ratio, $R_{6/5}$	Fissions attributable to U^{235}, F_5, at. %	Effective W^{184} fission cross section, σ_{eff}, b
1	0.001687	0.002091	0.212	1.63
2	.003727	.004075	.180	1.72
3	.001677	.001928	.132	2.13
4	.001677	.002020	.180	2.34
5	.001677	.002158	.252	1.97
Average				1.96±0.36

To determine the cadmium ratio for tungsten in the PBR, bare and cadmium covered cobalt and tungsten were irradiated in the RD-5 and RA-8 locations. The flux spectra in these locations bracket the conditions in HT-1. The cadmium ratio in the HT-1 location is thought to be just slightly higher than in RA-8. The nuclear environment in RA-8 and RD-5 is described in table II. A comparison of the cadmium ratios in table III shows a greater epithermal response in W^{184} than in cobalt. In RA-8 the epicadmium contribution to W^{184} activation was about 9 percent.

TABLE II. - NUCLEAR ENVIRONMENT OF

RA-8 AND RD-5 FACILITIES

[Reactor power, 60 MW.]

Facility	Gamma heating at 60 MW, W/g	Thermal flux, neutrons/ $(cm^2)(sec)$	Fast flux (>0.1 MeV), neutrons/ $(cm^2)(sec)$
RA-8	1.5 to 2.5	1×10^{14}	1.6×10^{13}
RD-5	0.4	8×10^{14}	5.7×10^{11}

TABLE III. - CADMIUM RATIOS OF

Co-59 AND W-184

Facility	Cobalt-59	Tungsten-184
RA-8	52.3	9.95
RD-5	185	32.8

We have examined the interferences of other radioactive tungsten isotopes and their daughters on the determination of W^{185}. The four radides that could conceivably contribute to the observed β activity are 130-day W^{181}, 24-hour W^{187}, 69-day W^{188}, and 17-hour Re^{188}.

Tungsten-181 decays by internal conversion. The efficiency for detection of the resultant X-rays in a proportional counter would be lower than for the W^{185} β's. The abundance of W^{180} in natural tungsten is only 0.14 percent; the abundance of W^{184} is 30.64 percent. Uncertainties in experimental attempts to determine the cross section of W^{180} are quite large but estimates based on the resonance integral indicate a value of about 2 barns (this cross section is based on information supplied by Brookhaven National Laboratory, May 1968). The anticipated contribution from W^{180} is less than 0.5 percent. For the short irradiations in these experiments the contribution would be about half of this because of the lower specific activity of W^{181}.

Tungsten-186 has a much higher cross section than W^{184} and a much higher specific activity. By waiting 20 days or 20 half-lives after irradiation there was not enough W^{187} to interfere with W^{185} activity determination. For long irradiations, however, the production of W^{188} by burnup of W^{187} must be considered. The capture cross section of W^{187} is about 90 barns.

The presence of W^{188} is detected by the gamma activity of the 17.0-hour Re^{188} daughter. No W^{188} activity was observed in irradiations of several hours. Estimates of the effect of W^{188} interference on the W^{185} activity determination show the W^{188}-Re^{188} contribution to total activity to be about 11 percent for a 1000-hour irradiation at 10^{14} neutrons per square centimeter per second. The relative W^{188} interference reaches a maximum after the W^{187} activity has reached equilibrium, then falls off with the burnup of W^{186}.

The reader is cautioned to check the response of W^{184} in the particular nuclear environment where its use is intended. We have investigated the response only for irradiations of 40 hours or less in a well-moderated neutron spectrum. The effect of W^{188} interference, which would be produced on long irradiations, has not been determined experimentally. The thermal cross section of W^{185}, though not considered to be large (Brookhaven National Laboratory, May 1968), is not known. We did not investigate the response of W^{184} to a fast-neutron spectrum.

SUMMARY AND CONCLUSIONS

The naturally occurring isotope, W^{184}, has been shown to be an effective thermal flux monitor for high-temperature (1100 to 3300 K), short-term irradiations of 40 hours or less.

The effective thermal cross section in this high-temperature application is 1.94±0.08 barns. The epicadmium contribution to W^{184} activation was determined to be about 9 percent. The radiochemical separation methods and counting techniques for W^{185} have been developed to allow an accurate determination of the weak β-activity. However, the accuracy of using W^{184} as a flux monitor depends heavily on the removal of other interferring radides during the W^{185} radiochemical separation.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, January 20, 1969,
122-29-05-08-22.

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